

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of : Confirmation No. 1456  
Eiichi KAJI et al. : Attorney Docket No. 2006\_1387A  
Serial No. 10/593,579 : Group Art Unit 1796  
Filed September 20, 2006 : Examiner C. Caixia LU

POLYMETHYLALUMINOXANE PREPARATION,  
METHOD OF PRODUCING THE SAME,  
POLYMERIZATION CATALYST, AND  
POLYMERIZATION METHOD OF OLEFINS

**DECLARATION UNDER 37 CFR 1.132**

Commissioner for Patents,

Alexandria, VA

Sir:

I, Eiichi Kaji, the undersigned, a citizen of Japan, residing at 1-2-502 Midori-machi Shunan city Yamaguchi, Japan, do hereby declare:

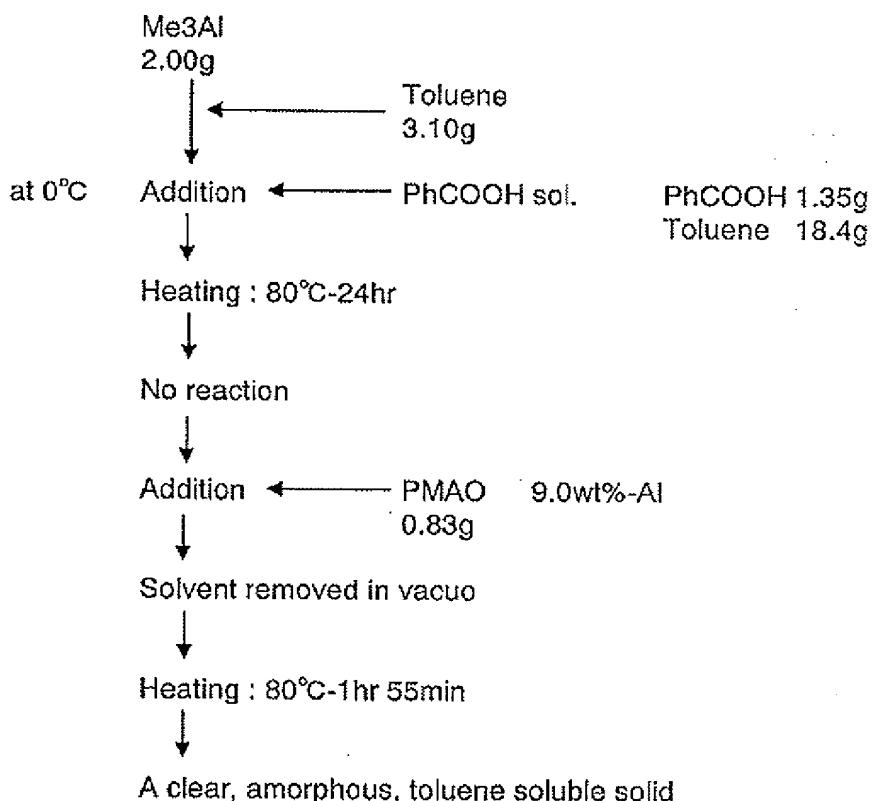
1. That I am a co-inventor of the above-identified application.
2. That I graduated from Japan Advanced Institute of Science and Technology on March, 1998 with a Dr. degree in Material Science.
3. (Relevant Work Experience)
4. (Relevant Publications, awards, or other distinguishing professional recognitions)
5. That in order to show the novelty and unobviousness of the subject matter of the above-identified application, I have under my control and direction conducted the following experiments. The particulars and results of the experiments are set forth hereinbelow.

### EXPERIMENTAL

Trace experiments were conducted according to Examples 2 and 4 of U.S. Patent No. 5,831,109 (Smith et al.) to confirm the actual results of Examples 2 and 4.

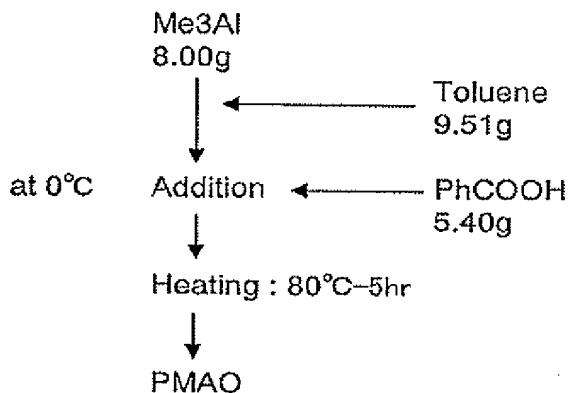
Illustrated below is the synthetic flows of Examples 2 and 4 of U.S. Patent No. 5,831,109.

#### Synthetic flow of Ex.2 of US 5,831,109



It is disclosed that in Example 2, when Me<sub>3</sub>Al and PhCOOH were mixed together and heated at 80°C for 24 hours, no reaction proceeded, and accordingly, a catalytic amount of PMAO was added to the mixture to complete the reaction. After the addition of PMAO, the solvent (toluene) was removed in vacuo to give a viscous liquid, which would be an improvement in this technique. It is also disclosed that the resulting PMAO was obtained as a solid material.

Synthetic flow of Ex. 4 of US 5,831,109



In Example 4,  $\text{Me}_3\text{Al}$  and  $\text{PhCOOH}$  were mixed together and heated at  $80^\circ\text{C}$  for 5 hours. This process did not involve the addition of catalytic PMAO and the removal of solvent, but gave PMAO.

Trace experiments were conducted according to the synthetic flows of Examples 2 and 4 of U.S. Patent No. 5,831,109, described above, in order to confirm the results, including the viscosity of obtained PMAO and stability thereof, until gel is formed.

Run numbers 1, 2, and 3 in Table 1 (below) show the results obtained through the trace experiments. Table 1 also includes the used amounts of starting materials, the molar ratio of  $\text{Al}/\text{O}$ , and the theoretical Al concentrations. Runs 1 and 2 correspond to Example 2, and Run 3 corresponds to Example 4.

Table 1

Run	Me <sub>3</sub> Al	Tol.	PhCOOH	Tol.	PMAO	Al/O*1	Al conc.*3
Ex.2*2	2g 27.75mmol	3.10g	1.85g 11.05mmol	18.4g	0.83g 2.77mmol (9wt%·Al)	1.26	3.25wt%
Ex.4*2	8g 110.99mmol	9.51g	5.40g 44.22mmol	None	None	1.25	13.50wt%
1	20.63g 282mmol	32.52g	13.61g 111.5mmol	188.04g	None	1.26	3.01wt%·Al
2	20.50g 280.22mmol	32.00g	13.52g 110.76mmol	189.21g	8.50g 28.02mmol (8.9wt%·Al)	1.26	3.18wt%·Al
3	80.05g 1095.00mmol	95.58g	53.36g	None	None	1.25	13.31wt%·Al

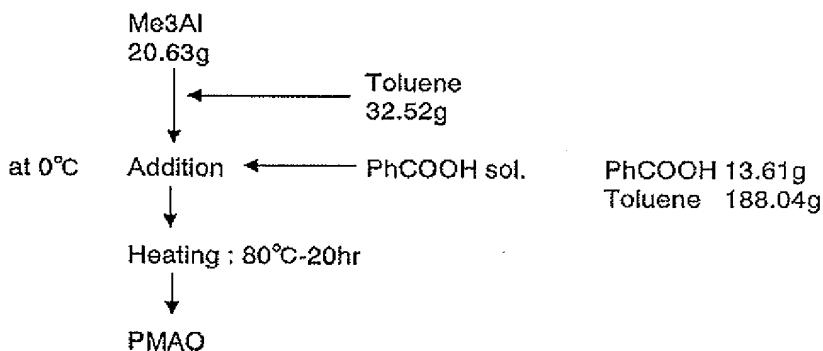
\*1 : Molar ratio of Me<sub>3</sub>Al to O derived from PhCOOH

\*2 : Data from Examples of U.S. 5,831,109

\*3 : Al concentration was calculated on the assumption that only CH<sub>4</sub> gas generated by the reaction was removed.

The following is a description of the synthesis of Run 1.

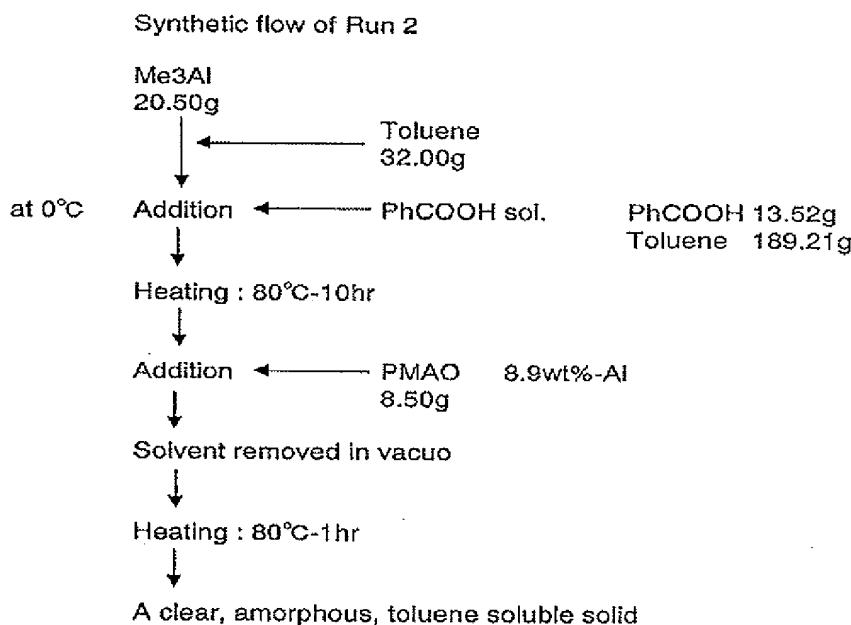
#### Synthetic flow of Run 1



The purpose of Run 1 was to confirm the synthesis according to Example 2. After the mixture was heated at 80°C for 20 hours, the reaction was completed. Since the PMAO formation was confirmed, catalytic PMAO was not added. The reaction mixture was collected and concentrated to prepare a 9wt% Al solution. Although the solution was a clear liquid immediately after the preparation, it showed deposition of gels on the following day (stored at 25°C).

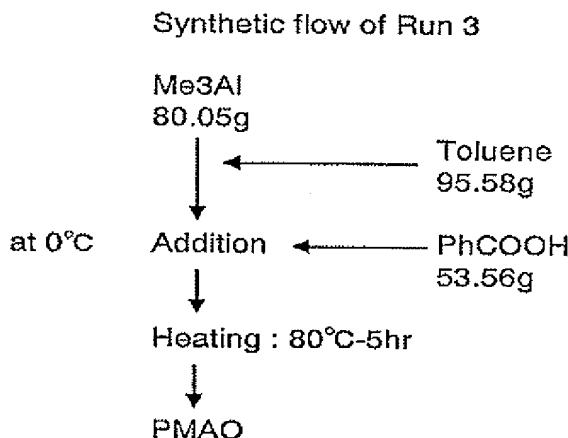
Accordingly, it was not possible to carry out its viscosity measurement.

This trace experiment was conducted for the purpose of the confirmation of the accurate synthesis according to Example 2. As described above, however, the PMAO formation was confirmed simply by heating at 80°C, without the addition of catalytic PMAO, meaning that the disclosed fact in Example 2 was not repeated. In addition to this, the 9wt% Al solution prepared from the obtained PMAO could be easily converted into gels, meaning that the stability thereof was extremely low. When part of the PMAO solution obtained in Run 1 was separately stored at 25°C without concentration treatment, it turned to gels on the following day even with the low concentration of 3wt% Al.



In Run 2, the reaction was completed without any catalytic PMAO added, although the addition of such catalytic PMAO is indicated as essential in Example 2. In order to confirm the effect of the PMAO solution added during the reaction, the mixture was heated for 10 hours, then the PMAO solution was added, and the solvent in the system was removed before the reaction was terminated. The resulting mixture was dissolved in toluene to prepare a 9wt% Al solution. The

resulting solution stored at 25°C showed gel formation on the following day, thus indicating poor storage stability.



The purpose of Run 3 was to confirm the synthesis according to Example 4. The reaction according to the trace experiment smoothly proceeds as described in Example 4 to give a viscous PMAO solution. The obtained PMAO solution was diluted with toluene to give a 9wt% Al solution. Then, the diluted solution and undiluted solution were examined for stability. These samples showed no gel formation on the following day (stored at 25°C) and their viscosities could be measured using a Ubbelohde viscometer. The results are shown in Table 2.

Table 2

	Sample	Viscosity
a)	Undiluted sample	8.21cP (at 32.0°C)
b)	Diluted sample	2.01cP (at 32.0°C)

Next, their storage stabilities at 25°C were examined. First the sample b) showed gel formation after 14 days. The sample a) had a high initial viscosity, and accordingly, was a highly viscous liquid. Although the change in viscosity was difficult to determine due to its high initial

viscosity, the observed viscosity gradually increased so that the solution became a syrup-like solution. The viscosity of the sample a) could not be determined after 20 days as the measurable range of the Ubbelohde viscometer is up to 20 cP. The addition of toluene to the sample a) showed generation of insoluble matters.

Smith et al. (U.S. Patent No. 5,831,109) disclose that PMAO obtained according to Examples 2 and 4 showed a high stability. In contrast to the disclosed results, the extremely low stability of PMAO obtained according to Examples 2 and 4 has now been confirmed, based on the trace experiments discussed above, when compared with the PMAO obtained according to the description of the subject application. It should be noted that the test results described in Smith et al. were significantly insufficient due to no disclosure with regard to the evaluation method, although they insisted the high storage stability of the obtained PMAO. In addition, it was confirmed that the trace experiment according to Example 2 could not be repeated.

In conclusion, the subject invention of the present application is not anticipated, or rendered obvious by the disclosure of Smith et al. (U.S. Patent No. 5,831,109).

I further declare that all statements made herein of my own knowledge are true and all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Date: June 16, 2009

Eiichi Kaji

(Signature of Declarant)